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International Intercalibration and Intercomparison Program Radon Daughter Measurements

Exercise at the Twilight Mine, Uravan, CO

By W. E. Cooper and R. F. Holub



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International Intercalibration and Intercomparison Program Radon Daughter Measurements

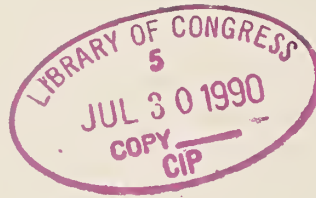
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UNIT OF MEASURE ABBREVIATIONS USED IN THIS REPORT

cm/s	centimeter per second	MeV	million electron volt
ft	foot	min	minute
h	hour	mm	millimeter
hp	horsepower	μm	micrometer
kBq/m ³	kilobecquerel per cubic meter	pCi/L	picocurie per liter
kPa	kilopascal	WL	working level
L/min	liter per minute		

INTERNATIONAL INTERCALIBRATION AND INTERCOMPARISON PROGRAM RADON DAUGHTER MEASUREMENTS

Exercise at the Twilight Mine, Uravan, CO

By W. E. Cooper¹ and R. F. Holub²

ABSTRACT

The International Intercalibration and Intercomparison Program (IIIP), consisting of several selected laboratories from four countries, held a radon progeny intercomparison measurement at the U.S. Bureau of Mines experimental Twilight Mine on September 12-14, 1988. Grab samples were taken at four different conditions of low and high radon progeny and condensation nuclei concentrations, respectively. The results showed good agreement among all seven participants. The coefficient of variation of all measurements was 4.7%; after minor corrections for flows and some systematic biases, it was reduced to 3.2%.

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INTRODUCTION

The accurate assessment of both occupational and general public exposure to radon and radon daughters is desirable to estimate their associated carcinogenic risk. For accurate exposure assessment, it is necessary to quantitatively evaluate the accuracy of different methods and equipment used to measure radon and radon daughter concentrations. In 1983, the Committee on Radiation Protection and Public Health (CRPPH) of the Organization for Economic Cooperation and Development, Nuclear Energy Agency (OECD/NEA) recognized this necessity and decided to set up an international program of intercalibration and intercomparison of equipment and techniques used for the monitoring of radon, thoron, and their short-lived daughters. From the beginning, this program was merged with a similar program conducted by the Commission of the European Communities (CEC). The primary purpose of the program was to quantitatively

assess measurement differences among international laboratories or groups to provide a forum for alleviating or reducing differences and inaccuracies. Because of the importance of accurate radon measurements in mining health and safety, the U.S. Bureau of Mines was one of the participating organizations from the United States.

The program was initially divided into three parts: intercalibration and intercomparison of radon measurements, intercalibration and intercomparison of radon daughter measurements (laboratory environment), and intercomparison of radon daughter measurements in real mine and dwelling conditions. The first two parts of the program have been completed and the results reported (1-2).³ This report summarizes the results of one of several intercomparisons performed at mines and dwellings that will be included in the report on the third part of the program.

ACKNOWLEDGMENTS

The authors acknowledge the assistance of T. H. Davis, electronics technician, and R. F. Drouillard, geophysicist, both of the Denver Research Center, in preparing the

mine, in performing the continuous measurements, and in organizing this exercise.

MEASUREMENT FACILITIES AND PROCEDURES

Radon daughter grab sample measurements for this intercomparison were taken in the controlled mine atmosphere of the Bureau's Twilight Mine, located about 10 miles northwest of Uravan, CO. The mine is a previously operating uranium-vanadium mine, which was acquired by the Bureau to conduct in-house and contract research. The ore zone consists of a well-sorted, fine-grained, highly fractured sandstone with abundant carbonaceous material. A plan view map of the mine configuration and facilities is included as figure 1. The mine and its facilities are described in detail elsewhere (3). The measurements were done in the air-cleaning test area shown in figure 1.

The ventilation system of the mine consists of twin 20-hp primary fans (back to back) located at the main exhaust portal (portal B) and a two-stage, 7-1/2-hp each, secondary fan located at bulkhead B of the North Loop (fig. 1). The secondary fan automatically turns on in the event of power failure. The primary fan ran continuously throughout the intercomparison, while the secondary fan was used only as needed to provide recirculation around the North Loop, thereby increasing radon daughter concentrations in the measurement area. A small diesel engine was situated at drift IL5 and used as required to entrain diesel

pollutants into the mine air. Throughout the intercomparison, continuous measurements of radon, radon daughters, barometric pressure, and relative humidity were taken to evaluate factors that may affect radon daughter concentrations. All of these measurements, except barometric pressure, were taken in the measurement area shown in figure 1. Barometric pressure was measured outside the mine near the instrument trailer. Details about the monitoring equipment are given in reference 3.

The intercomparison participants and their equipment and procedures are listed in tables 1 and 2. Grab samples were taken under four conditions:

1. Low radon daughter concentration (0.22 to 0.50 WL) with no diesel pollutants;
2. Low radon daughter concentration (0.31 to 0.33 WL) with diesel pollutants;
3. High radon daughter concentration (0.86 to 0.94 WL) with no diesel pollutants;

³Italic numbers in parentheses refer to items in the list of references preceding the appendix.

4. High radon daughter concentration (0.73 to 0.92 WL) with diesel pollutants.

Table 1.—Participants¹

The ranges of radon daughter concentrations presented are the ranges of the average concentration obtained by the participants. The participants' filter holders were positioned near the middle of the mine airway with the filter faces directed toward the mine airflow. Mine air velocities (about 50 cm/s) in the measurement area were higher than the pump flow rate air velocities at the filter face. The filter holders were kept within a few centimeters of each other to help minimize effects due to measurement of different air.

	Abbreviation
Atomic Energy Control Board	AECB
Australian Radiation Laboratory	ARL
Environmental Measurements Laboratory, U.S. Department of Energy	EML
Department of Energy, Mines, and Resources, CANMET	EMR
Mine Safety and Health Administration, U.S. Department of Labor	MSHA
Bureau of Mines, U.S. Department of the Interior	USBM
University of Salzburg	U.S.

¹See appendix for more detailed listing.

Table 2.—Participant equipment and procedures

Participant	Detector		Counter efficiency	Filter		Pump flow rate, L/min	Method
	Type	Manufacturer		Type	Diam, mm	Pore size, μ m	
AECB . . .	ZnS Trimet.	NURAD . . .	0.435 .440	Millipore membrane AA.	25	0.8	5-min modified Tsivoglou.
ARL	ZnS drawer assembly.	ARL444	Gelman membrane GA-4.	25	.8	Do.
EML	ZnS Th-29-B TD-19.	ELM470	Reeve angel glass fiber, 934AH.	50	NAp	Do.
EMR . . .	ZnS Trimet.	NURAD467	Millipore membrane AA.	25	.8	Do.
MSHA . .	ZnS radon-flask detector.	Ludlum . . .	^{1,2} .440	Gelman glass fiber A/E.	25	NAp	Do.
			³ .476				
			⁴ .488	Millipore membrane AA.	25	.8	
USBM do. do.477	. . do. . . .	25	.8	Do.
U.S. . . .	Silicon-diode.	Pylon20	Pylon	25	NAp	Alpha-spectroscopy (RaA-RaC').

NAp Not applicable.

¹Filter self-absorption included.

²Sept. 12, 1988.

³Sept. 13, 1988.

⁴Sept. 14, 1988.

NOTE.—Reference to specific products does not imply endorsement by the U.S. Bureau of Mines.

IR1 Power center
 IR2 Galf-cart charging system
 IL1 Protective clothing area
 IL2 Storage area
 IL3 Charging center

Site	Width, ft	Length, ft
IR3	10	25
IR5		
IR5-1	10	25
IR5-2	10	30
IR6	8	40
IR7	10	25
IR8	10	55
IL4	15	50
IL5	20	50
IL6		
IL6-1	10	20
IL6-2	20	20
IL7	30	65
IL8	30	25
IL9	15	
IL10	10	10
2R1	15	35
2R2	10	7
2R3	10	25
2R4	10	27
2R5	10	35
2R6	10	55
3R1	10	15
3R2	10	5
3R3	10	5
3R4	20	15
3L1	10	5
3L2		
3L2-1	10	5
3L2-2	10	30
3L2-3	10	15
3L2-4	10	5
3L3	8	35
3L4	7	115
3L5	18	20
3L6		
3L6-1	20	18
3L6-2	20	10
3L6-3	10	35
3L6-4	10	30
3L6-5	10	30
3L6-6	10	10
3L7	20	20

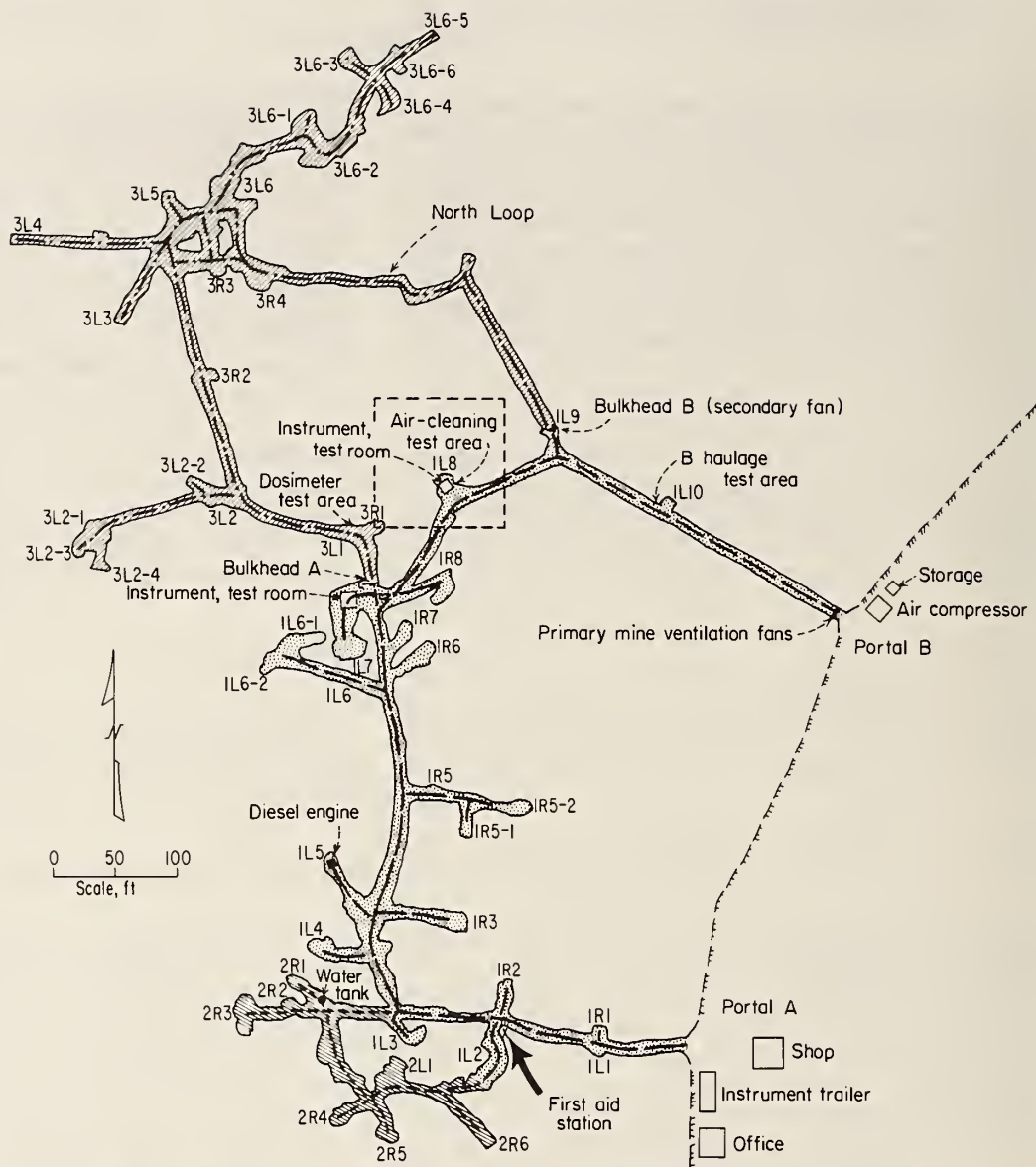


Figure 1.—Plan map of the Twilight Mine. North Loop is defined by bulkheads A and B.

RESULTS AND DISCUSSION

The results of the continuous measurements of radon, radon daughters, barometric pressure, and relative humidity are presented in figures 2-4. Also shown on these figures are the time periods for which the diesel engine was operating and the bulkhead B secondary fan (North Loop fan) was on. The relative humidity was high (60% to 70%) on September 12, 1988, because of rain occurring outside the mine. The continuous radon daughter concentration measurements lag behind the radon concentration measurements about 1 h because of the detection methods utilized. As expected, the ratio of the radon daughter concentration over the radon concentration increased with the entrainment of diesel pollutants. These figures also show that barometric pressure inversely affected the radon concentration, as reported previously (4).

The reported results of the individual radon daughter concentration measurements for three consecutive days are presented in table 3. Also included are the calculated average concentration and coefficient of variation for each sample time. These results showed average coefficients of variation of 13%, 10%, and 15%, respectively, for RaA, RaB, and RaC at low concentrations (0.22 to 0.50 WL). These low concentration results corresponded to average concentrations of 89, 33, and 22 pCi/L, respectively, for RaA, RaB, and RaC. At high concentrations (0.73 to 0.94 WL), September 14, the results corresponded to average concentrations of 170, 91, and 64 pCi/L and coefficients of variation of 11%, 6.7%, and 8.2%, respectively, for RaA, RaB, and RaC. Lower coefficients of variation at the high concentrations were expected because of a reduction in the statistical counting uncertainty.

The reported radon daughter concentrations are presented in table 4. Also listed in the table are the average concentrations for each sample time and the coefficients of variation for the sampling results at each time. The coefficients of variation ranged from 11.4% to 2.1%, with an overall average coefficient of variation of 6.0%.

The reported radon daughter sampling results for each of the participants was analyzed further by calculating the ratio of the reported working level concentration and the average concentration for each sample time. These calculated results, along with the average ratio and standard deviation for each participant, are listed in table 5. The average ratio provides an indication of the average percent that the participants' results were above or below the average concentration. The results showed an average ratio range of 0.957 to 1.046. The standard deviation provides an indication of the variability of the participants' sampling results. Some uncertainty in determining the actual working level would also be included in the listed standard deviations because the average concentration was used to estimate the actual working level. The average standard deviation for the participants (the last column in table 5) was only 4.7%. This low standard

deviation indicates that the measurement uncertainty for each of the participants was relatively constant. It also indicates that the differences in average ratio among the participants are probably largely due to systematic bias.

The amount of systematic bias due to computational differences in converting individual radon daughter concentrations to working levels was analyzed by converting all of the reported individual daughter concentrations to working levels using a constant conversion factor and comparing these results with the reported working level concentrations. The ratio of the calculated working level and the reported working level was computed and the average ratio for each participant determined. The results of these calculations are presented in table 6.

From the table 6 results, the Environmental Measurements Laboratory (EML) and the Mine Safety and Health Administration (MSHA) showed some systematic bias when converting individual radon daughter concentrations to working levels. These systematic biases were calculated at about 1% for EML and 2% for MSHA. The bias for MSHA resulted from using a conversion factor of $1.28\text{E}5 \text{ MeV} = 1 \text{ WL}$ instead of $1.3\text{E}5 \text{ MeV} = 1 \text{ WL}$. For EML, the indicated bias is probably due to two factors: (1) the use of half-lives of 3.11 min and 19.9 min, respectively, for RaA and RaC instead of 3.05 min and 19.7 min, and (2) reporting the individual radon daughter concentrations to only two significant digits, while the other participants reported their results to three significant digits. The lower number of significant digits resulted in a higher standard deviation for the ratio than those obtained for the other participants. This higher standard deviation would result in a higher uncertainty for EML's average ratio. These estimated systematic biases should result in their measurements being above the average results by the appropriate percentages.

It was noticed that the Department of Energy, Mines, and Resources (EMR), MSHA, and the Atomic Energy Control Board (AECB) used the same pump calibration device at the minesite and all three participants' results were above the average. This prompted a further analysis of their pump calibration device in the laboratory. The pump calibration device used was a Gilibrator, similar to the Buck Calibrator recently tested by MSHA (5). The Buck Calibrator showed a systematic bias of 1.4% at a flow rate of about 2.0 L/min (5). A laboratory comparison of the Gilibrator against a Brooks flowmeter at 4.0 L/min showed a systematic bias of about 2.5%. This indicates that the results of the three participants are probably systematically biased about 2.5% high because of an inaccurate pump flow rate calibration at the time of the intercomparison. However, as was apparent during the last intercomparison in France, in June 1989, the problem of measuring flow has not been satisfactorily resolved.

Table 3.—Radon daughter concentrations, picocuries per liter

Date and participant	Time: 14:01			Time: 15:15			Time: 16:09		
	RaA	RaB	RaC	RaA	RaB	RaC	RaA	RaB	RaC
Sept. 12, 1988:									
AECB	95.7	35.7	17.6	111.3	32.1	13.9	82.2	31.4	21.8
ARL	84.9	32.1	22.9	105.4	29.8	16.5	88.6	34.9	17.8
EML	NA	NA	NA	89.1	30.2	17.0	81.0	31.6	22.1
EMR	NA	NA	NA	NA	NA	NA	NA	NA	NA
MSHA	69.3	31.1	22.8	83.6	27.7	18.9	78.2	30.5	20.4
USBM	74.7	26.4	26.2	93.7	28.6	16.2	94.5	34.3	14.8
Average	81.15	31.33	22.38	96.61	29.70	16.51	84.90	32.54	19.39
COV %	14.4	12.2	15.9	11.9	5.7	10.8	7.8	6.0	15.9
	Time: 9:00			Time: 10:00			Time: 11:00		
	RaA	RaB	RaC	RaA	RaB	RaC	RaA	RaB	RaC
Sept. 13, 1988:									
AECB	NA	NA	NA	51.6	19.2	20.0	68.4	32.0	31.3
ARL	60.1	18.6	16.2	62.3	22.3	13.0	84.9	38.7	24.9
EML	NA	NA	NA	62.1	20.0	13.0	72.9	37.8	24.3
EMR	61.7	22.5	15.8	57.2	24.8	15.6	99.3	40.3	22.4
MSHA	79.9	22.6	17.3	46.8	18.7	22.3	79.9	40.9	31.4
USBM	NA	NA	NA	60.3	17.9	12.6	70.8	34.5	26.4
Average	67.23	21.23	16.43	56.71	20.48	16.08	79.37	37.37	26.79
COV %	16.4	10.7	4.7	11.1	12.7	25.7	14.5	9.2	14.1
	Time: 12:00			Time: 13:00			Time: 14:00		
	RaA	RaB	RaC	RaA	RaB	RaC	RaA	RaB	RaC
AECB	77.6	36.2	21.1	96.5	28.1	20.5	112.9	40.4	23.6
ARL	72.0	25.5	19.4	86.7	30.1	17.8	121.0	35.6	21.4
EML	56.7	27.5	28.4	72.9	29.4	17.8	108.0	38.6	18.4
EMR	103.7	38.3	21.2	116.2	30.0	18.8	102.4	43.9	27.7
MSHA	88.4	37.2	24.2	78.5	21.5	26.5	108.9	37.6	27.4
USBM	69.7	29.8	23.6	81.3	32.8	16.1	124.0	37.0	20.5
Average	78.2	32.42	22.97	88.68	28.66	19.59	112.87	37.36	23.16
COV %	20.9	16.9	13.8	17.7	13.3	18.8	7.3	5.4	16.4
	Time: 15:00			Time: 16:00					
	RaA	RaB	RaC	RaA	RaB	RaC			
AECB	143.0	52.4	26.8	112.9	41.7	29.7			
ARL	148.0	45.4	28.5	114.0	39.1	28.8			
EML	121.5	43.2	27.0	116.1	45.9	21.6			
EMR	169.6	54.5	25.2	144.8	51.8	26.8			
MSHA	157.1	50.6	31.8	107.2	42.7	31.8			
USBM	137.9	45.7	33.2	124.6	43.7	29.8			
Average	146.18	48.64	28.76	119.94	44.15	28.08			
COV %	11.3	9.3	10.8	11.2	9.9	12.7			

See notes at end of table

Table 5.—Ratio of reported working levels over average working levels

Participant	Sept. 12, 1988			Sept. 13, 1988							
	14:01	15:15	16:09	9:00	10:00	11:00	12:00	13:00	14:00	15:00	16:00
AECB	1.049	1.043	0.993	NA	1.005	0.943	1.028	1.031	1.051	1.017	0.963
ARL	1.024	1.024	1.027	0.907	1.014	1.018	.835	1.005	.997	.975	.935
EML	NA	.992	1.006	NA	.961	.962	.925	.956	.977	.900	.972
EMR	NA	NA	NA	.990	1.090	1.056	1.151	1.115	.997	1.086	1.133
MSHA988	.977	.969	1.103	1.028	1.115	1.133	.950	1.059	1.082	.996
USBM939	.964	1.006	NA	.902	.932	.937	1.005	1.007	.985	1.014
U.S.	NA	NA	NA	NA	NA	.973	.991	.940	.912	.955	.987
SD048	.033	.021	.098	.064	.067	.114	.061	.049	.067	.064
	Sept. 14, 1988							Av	SD		
	9:00	10:00	11:00	12:00	13:00	14:01	14:59	ratio			
AECB	1.139	1.083	1.064	1.099	1.046	1.036	1.041	1.037	0.047		
ARL	1.061	.989	1.000	.979	.964	.979	.941	.982	.053		
EML980	.927	.933	.959	.926	.988	.945	.957	.029		
EMR	1.045	.976	1.002	.978	.999	1.008	1.061	1.046	.058		
MSHA	1.019	1.049	1.044	1.003	1.020	1.044	1.022	1.033	.051		
USBM894	.986	.991	1.012	.966	.944	.989	.969	.038		
U.S.862	.989	.967	.970	1.079	NA	NA	.966	.054		
SD097	.051	.044	.047	.053	.038	.050	NAp	NAp		

NA Not available.
NAp Not applicable.
SD Standard deviation.

Table 6.—Ratio of calculated working levels using reported radon daughter concentrations over reported working levels

Participant	Sept. 12, 1988			Sept. 13, 1988							
	14:01	15:15	16:09	9:00	10:00	11:00	12:00	13:00	14:00	15:00	16:00
AECB	1.001	1.002	1.001	NA	1.001	1.000	1.003	1.002	1.001	1.001	1.001
ARL996	.995	.996	.994	.995	.997	.996	.995	.993	.994	.996
EML	NA	.986	.992	NA	.994	1.002	.990	.986	.989	.980	.980
EMR	NA	NA	NA	.994	.996	.996	.995	.994	.995	.995	.994
MSHA967	.965	.982	.987	.984	.983	.984	.986	.984	.984	.984
USBM999	.994	.993	NA	.990	1.002	1.000	1.001	1.000	1.002	1.001
	Sept. 14, 1988							Av	SD		
	9:00	10:00	11:00	12:00	13:00	14:01	14:59	ratio			
AECB	1.001	1.001	1.001	1.001	1.001	1.001	1.001	1.001	0.001		
ARL998	.997	.997	.998	.998	NA	NA	.996	.002		
EML974	.999	.987	.984	.982	1.005	.983	.988	.008		
EMR997	.997	.997	.997	.996	.996	.994	.996	.001		
MSHA984	.984	.984	.983	.984	.984	.984	.982	.006		
USBM	1.001	1.001	1.001	1.000	1.001	1.002	1.002	.999	.004		

NA Not available.
SD Standard deviation.

NOTE.—WL = (RaA × 0.0010287) + (RaB × 0.00507745) + (RaC × 0.0037323), where RaA, RaB, and RaC are concentrations in picocuries per liter.

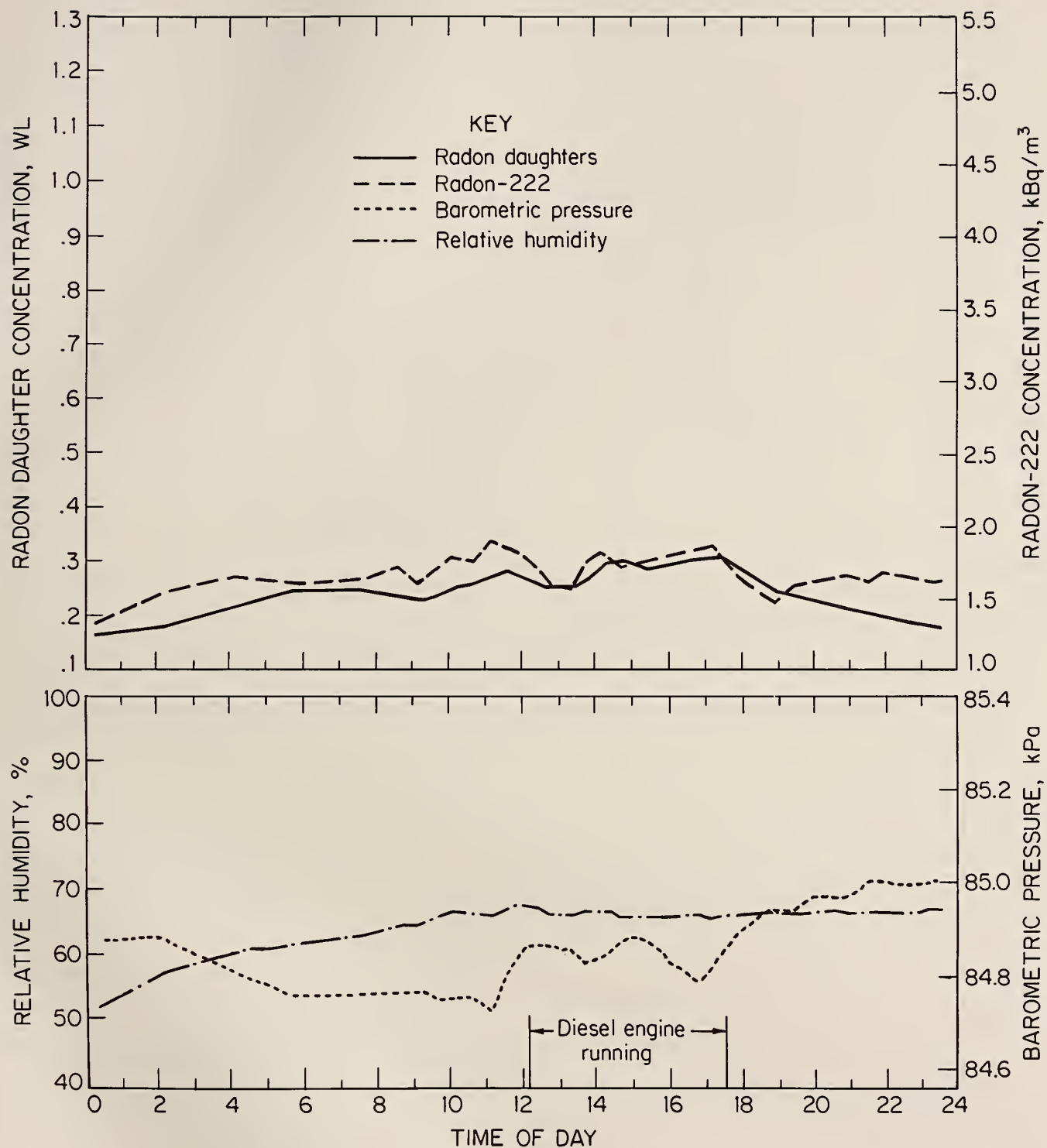


Figure 2.—Total radon daughters, radon, barometric pressure, and relative humidity as functions of time, September 12, 1988.

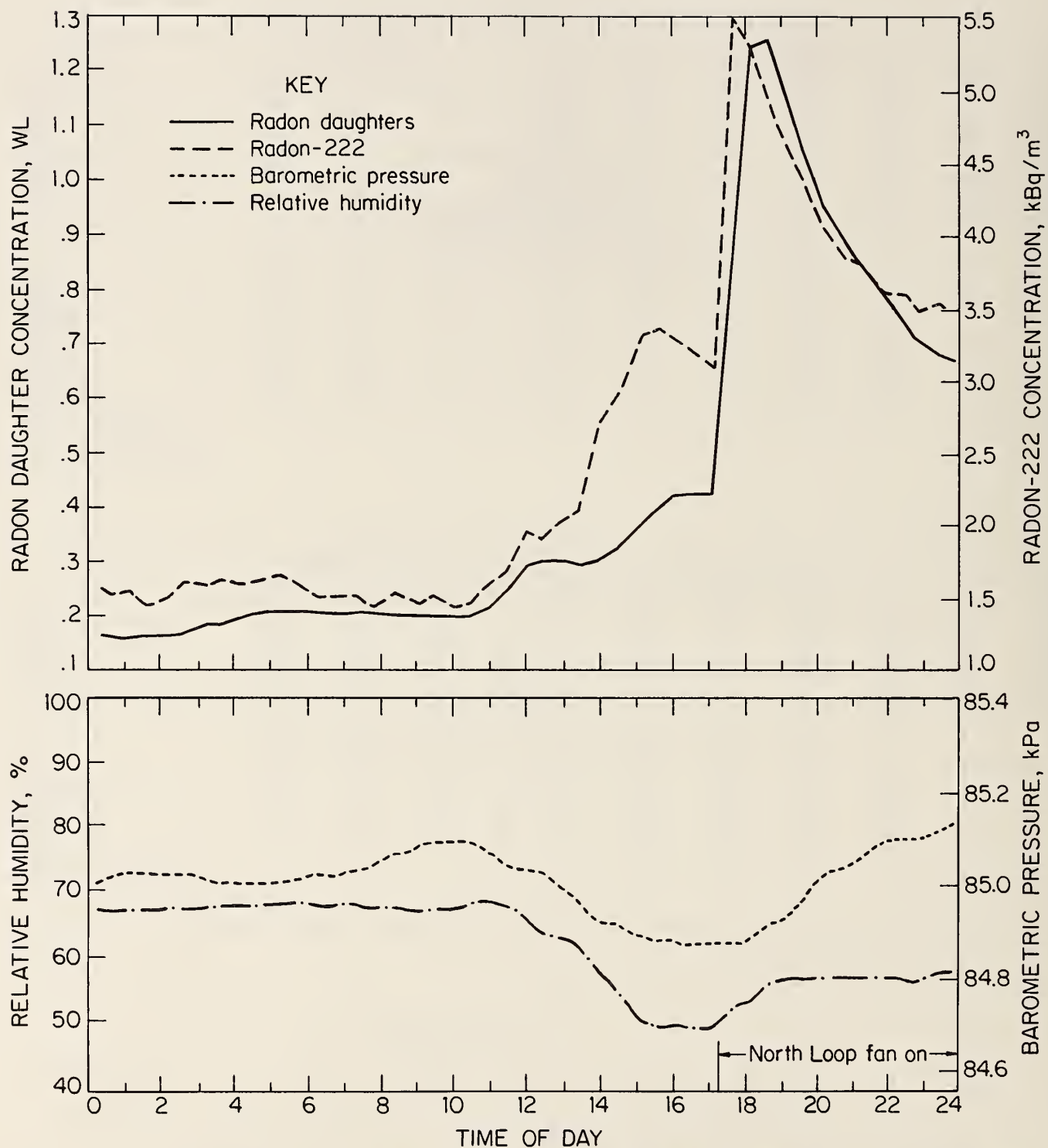


Figure 3.-Total radon daughters, radon, barometric pressure, and relative humidity as functions of time, September 13, 1988.

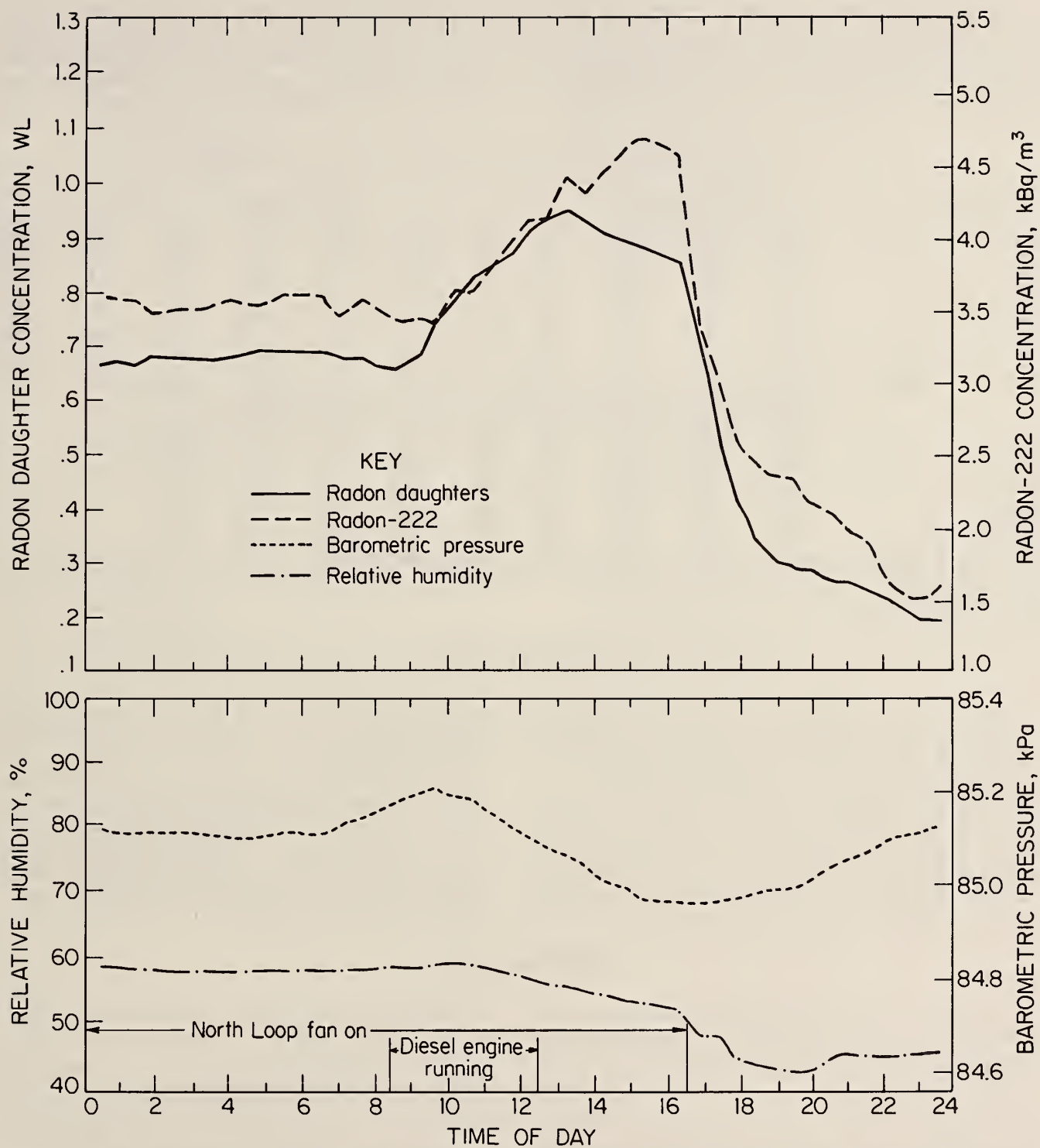


Figure 4.-Total radon daughters, radon, barometric pressure, and relative humidity as functions of time, September 14, 1988.

The pump flow rates for the affected three participants were adjusted for the probable systematic bias due to pump miscalibration (2.5%), and the results are presented in table 7. The pump flow rate corrections indicate that the maximum average percentage that a participant's

results were above or below the average concentration was only 3.2%. These results appear quite good considering these were field measurements and some difference would be expected because of measurement of different air and statistical counting uncertainties in the methods.

Table 7.—Ratios of flow-corrected working levels over average working levels

Participant	Sept. 12, 1988			Sept. 13, 1988							
	14:01	15:15	16:09	9:00	10:00	11:00	12:00	13:00	14:00	15:00	16:00
AECB	1.036	1.028	0.978	NA	0.993	0.930	1.014	1.016	1.036	1.003	0.950
ARL	1.037	1.034	1.037	.923	1.027	1.030	.845	1.016	1.008	.986	.945
EML	NA	1.002	1.015	NA	.973	.973	.936	.967	.987	.910	.983
EMR	NA	NA	NA	.983	1.077	1.042	1.136	1.099	.983	1.071	1.117
MSHA	.976	.962	.954	1.094	1.015	1.100	1.118	.936	1.044	1.067	.982
USBM	.951	.974	1.015	NA	.914	.943	.948	1.016	1.018	.996	1.025
U.S.	NA	NA	NA	NA	NA	.983	1.003	.950	.922	.966	.998
SD	.043	.032	.033	.087	.055	.060	.103	.055	.041	.056	.058
	Sept. 14, 1988							Av	SD		
	9:00	10:00	11:00	12:00	13:00	14:01	14:59	ratio			
AECB	1.123	1.068	1.049	1.084	1.032	1.024	1.029	1.023	0.046		
ARL	1.073	1.000	1.011	.990	.974	.992	.953	.993	.053		
EML	.991	.938	.944	.970	.937	1.001	.958	.968	.029		
EMR	1.031	.963	.988	.964	.985	.996	1.049	1.032	.057		
MSHA	1.005	1.034	1.030	.989	1.006	1.032	1.009	1.020	.051		
USBM	.904	.997	1.001	1.023	.977	.956	1.002	.980	.038		
U.S.	.871	1.000	.978	.981	1.091	NA	NA	.977	.055		
SD	.089	.043	.035	.042	.050	.027	.038	NAp	NAp		

NA Not available.
NAp Not applicable.
SD Standard deviation.

CONCLUSIONS

The maximum average percentage that a participant's results were above or below the average concentration for each sample time was only 4.7%. After pump flow rate correction for three of the participants, which was necessary because of an inaccurate pump calibration device, this maximum reduced to 3.2%. Systematic biases of about 1% and 2%, respectively, in two of the participants' results were due to differences in converting individual daughter concentrations to working levels. The results of the intercomparison appeared to be reasonably good when

consideration is given to the statistical counting uncertainties involved and the possibility that the participants measured different air.

The intercomparison was beneficial in identifying measurement errors due to inaccurate pump calibration and different procedures for converting daughter concentrations to working levels. Additional intercomparisons should be periodically performed to assist laboratories in identifying measurement errors and helping to assure accurate exposure assessments.

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